

PULSED ELECTRON EMISSION FROM PLZT CERAMICS

H. Gundel¹⁾, J. Handerek²⁾, H. Riege¹⁾, E.J.N. Wilson¹⁾
and K. Zioutas³⁾

- 1) CERN, Geneva, Switzerland.
- 2) University of Silesia, Institute of Physics, Katowice, Poland.
- 3) University of Thessaloniki, Nuclear and Elementary Particle Physics Section, Thessaloniki, Greece.

ABSTRACT

New experimental results on electron emission from PLZT ceramics of composition 2/95/5, subjected to rectangular high-voltage pulses are reported. The dependence of the emitted electron charge on the electric field strength and temperature was investigated. The energy of the emitted electrons was estimated by applying a decelerating potential to an auxiliary grid electrode. Emitted current densities of several A/cm² and emitted charges up to $\sim 1 \mu\text{C}/\text{cm}^2$ ($\sim 10^{13}$ electrons/cm²) were observed.

Paper presented at the 7th International Meeting on Ferroelectricity, Saarbrücken, FRG, 28.8-1.9.1989 and to be published in Ferroelectrics.

Geneva, Switzerland
August 1989

PULSED ELECTRON EMISSION FROM PLZT CERAMICS

H. Gundel¹⁾, J. Handerek²⁾, H. Riege¹⁾, E.J.N. Wilson¹⁾ and K. Zioutas³⁾

1) CERN, Geneva, Switzerland.

2) University of Silesia, Institute of Physics, Katowice, Poland.

3) University of Thessaloniki, Nuclear and Elementary Particle Physics Section, Thessaloniki, Greece.

Abstract New experimental results on electron emission from PLZT ceramics of composition 2/95/5, subjected to rectangular high-voltage pulses are reported. The dependence of the emitted electron charge on the electric field strength and temperature was investigated. The energy of the emitted electrons was estimated by applying a decelerating potential to an auxiliary grid electrode. Emitted current densities of several A/cm² and emitted charges up to $\sim 1 \mu\text{C}/\text{cm}^2$ ($\sim 10^{13}$ electrons/cm²) were observed.

1. INTRODUCTION

Electron emission from barium titanate, from some chemically similar materials (LiNbO₃, LiTaO₃, etc.) and from a few other ferroelectrics (such as TGS, KDP, RS) has been investigated during the last 30 years (see review in Ref. 1). A variety of procedures—thermal, optical, and mechanical, or any of their combinations—were used to cause fast polarization changes and electron emission from a ferroelectric surface. The emitted current density was, however, at most 10^{-9} A/cm² and was much less in most cases.²⁻⁸

Investigating the electron emission from Pb(Zr,Ti)O₃ (PZT) and (Pb,La)(Zr,Ti)O₃ (PLZT) solid solutions we have obtained current and charge densities which are orders of magnitude greater.⁹ Additional results, obtained recently, are reported in the present paper.

2. EXPERIMENTAL TECHNIQUE

The experiments have been performed with a new PLZT-2/95/5 ceramic material having a considerably higher antiferroelectric (AFE)–ferroelectric (FE) phase transition temperature in comparison with the ceramics investigated earlier.⁹ In the former case⁹ 2% mol of La₂O₃ were added to a PZT-95/5 solid solution, whereas the present PLZT-2/95/5 ceramic was sintered according to the formula (Pb_{0.98}, La_{0.02})(Zr_{0.95}, Ti_{0.05})O₃. The considerable change of the AFE–FE phase transition temperature as a result of such a small difference in composition follows from PZT and PLZT phase diagrams.^{10,11} The AFE–FE phase boundary is an abruptly changing function near a Ti-content of 5% mol. The PLZT disks of 1 mm thickness and 17 mm diameter were polished and covered with evaporated gold electrodes ($\varnothing \sim 10$ mm), solid (SE) on one side and in the form of a striped electrode (GE) on the opposite face. The samples were pre-poled by a d.c. field (~ 2 kV/cm), with the negative potential on the GE electrode, at temperatures higher than the AFE–FE phase transition temperature and by cooling down under this field.

The electrical circuits and the mechanical set-up, with the measuring and auxiliary electrodes, were described in the earlier article.⁹ The striped electrode (GE) of the ferroelectric sample, mounted in a vacuum-tight support, was exposed to the vacuum ($p \sim 10^{-6}$ mbar). To cause electron emission negative HV pulses were applied to the SE sample electrode whereas GE was grounded.

There was considerable pulse-to-pulse scatter in the current and charge measurements, which decreased in general as the applied voltage and temperature were increased. The experimental data on the emitted charge, given below, represent the average over 10 randomly chosen shots. The charge, measured on the Faraday cup, has been multiplied by a factor of 140 to allow for the transparency of the grids and for the area of the bare part of the sample surface with the striped electrode.

3. EXPERIMENTAL RESULTS

The temperature dependence of the emitted electron charge density at constant electric field strength ($E = 25$ kV/cm) is shown in Fig. 1. The emitted charge measurements were performed as the sample was slowly heated across the AFE-FE phase transition temperature. A steep rise

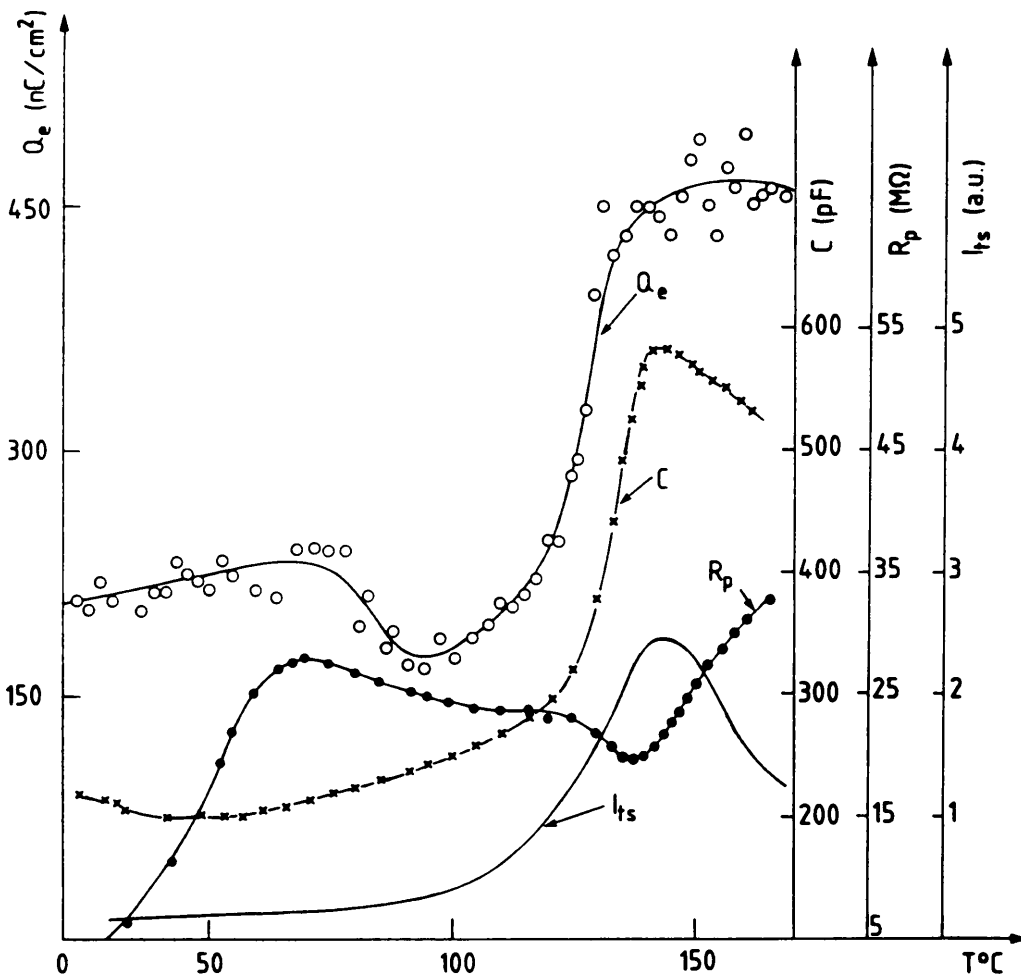


FIGURE 1 Emitted electron charge density (Q_e), capacity (C), resistance (R_p), and thermostimulated current (I_{ts}) as a function of temperature in the vicinity of the AFE-FE phase transition.

PULSED ELECTRON EMISSION FROM PLZT CERAMICS

of the emitted charge density takes place near this phase transition. There is an evident correlation between the maximum of the emitted charge and the temperature dependences on sample capacity (C) and resistance (R_p), which were measured with an a.c. field of 1 kHz. The broad local maximum in the $C(T)$ curve characterizes the AFE-FE diffuse phase transition (DPT). A similar but much more distinct maximum occurs at the FE-PE phase transition at 235°C (not shown in Fig. 1). The temperature variation of the thermostimulated current (I_{ts}), recorded during heating of the previously pre-poled sample, is also shown in Fig. 1. The maximum in the curve $I_{ts}(T)$ also coincides with the AFE-FE phase transition.

The influence of the electric field strength on the emitted charge density, studied at a few constant temperatures, is shown in Fig. 2. The emission process starts at a threshold field of

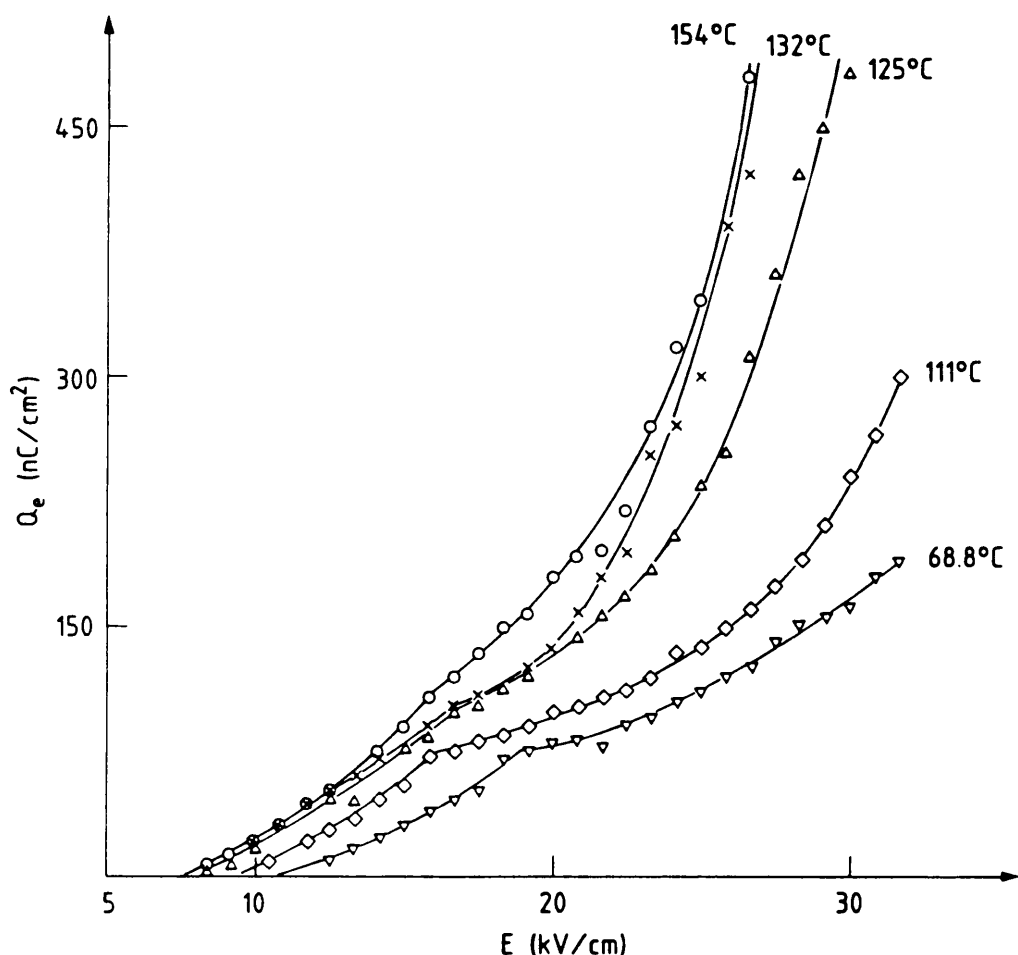


FIGURE 2 The emitted electron charge density as a function of the applied electric field strength at different temperatures near the AFE-FE phase transition.

about 7 to 10 kV/cm, which is decreasing with increasing temperature. The emitted current and charge increase strongly as higher electric field is applied. The electron emission is also strongly enhanced by a temperature increase in the vicinity of the AFE-FE DPT, which was already visible in Fig. 1. The curve $Q_e(E)_T$ (Fig. 2) is clearly divided into two regions of different slope.

We limited the applied field amplitude in order to prevent breakdown, to avoid perforation of the sample, and to ensure the use of the same sample for all measurements.

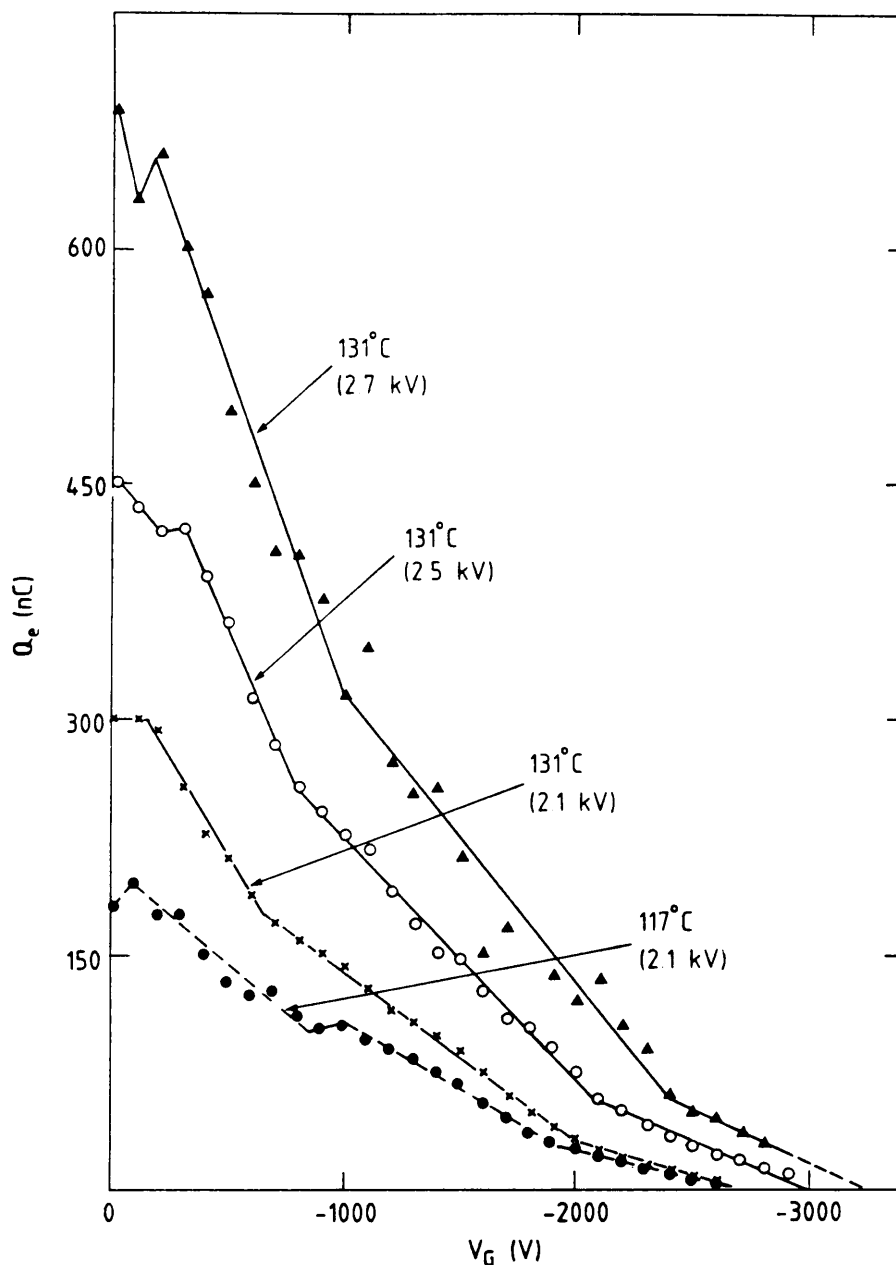


FIGURE 3 The charge measured on the Faraday cup as a function of decelerating potential applied to the auxiliary grid. The three solid curves are obtained at constant temperature ($T = 131^\circ\text{C}$) with different voltages applied to the sample. The lowest solid line and the dashed one refer to two different temperatures, but to the same applied voltage.

The charge emitted at a constant temperature, with three applied voltages as parameter, is shown in Fig. 3 as a function of decelerating voltage applied to the auxiliary grid. At least two linear regimes are visible in the curve $Q_e(V_G)_{T,E}$. This allows us to determine approximately the energy of the major part of the emitted electrons. This energy is of the order of a few keV, which corresponds to the applied voltage on the sample. The energy of the emitted electrons seems to be independent of the temperature of the sample.

PULSED ELECTRON EMISSION FROM PLZT CERAMICS

4. DISCUSSION

In the region of diffuse phase transition the fast reversal of spontaneous polarization P_s in existing FE domains and the appearance of new FE domains in the AFE matrix due to the AFE-FE phase transition shift under electric field both play an important role in the process of copious electron emission from Zr-rich PLZT ceramics. The changes in the electron states of the defects concentrated in the surface layers as F-centres (vacancies in the oxygen sublattice) in the previously pre-poled sample are equally important. If the temperature is low enough, the majority of the F-centres are filled by two electrons forming F_2 -centres. The electrons from the F_2 -centres (behaving as donors) are thermally liberated with increasing temperature. Such processes give rise to the thermostimulated current which was investigated often in the past in dielectric and semiconducting materials. It has been shown before¹² that in the vicinity of the intermediate ferroelectric phase of $PbZrO_3$ the thermally stimulated electron liberation process is increased under the influence of the electric field associated with P_s gradually appearing during AFE-FE DPT. The P_s vectors are partially oriented, at least in the surface layers, by the weak electric field formed by the resultant space-charge polarization of the pre-poled sample. The local maximum of the thermostimulated current and the local minimum of the resistance (R_p) in the DPT region can be explained in the same way (Fig. 1).

We think that the rapid increase of the emitted electron charge density in the AFE-FE phase transition region (Fig. 1) is of the same origin. Fast and strong P_s changes due to the applied HV pulses lead to copious electron liberation mainly by field generation. Afterwards, they are emitted from the bare part of the sample surface. The shift of the AFE-FE phase transition temperature towards lower temperatures when an electric field is applied is particularly strong in the case of materials containing more than 3% mol Ti.¹¹

All processes of P_s change and electron generation depend strongly on the electric field strength and temperature. This argumentation allows us to understand, in a qualitative way, the behaviour of $Q_e(T)_E$ and $Q_e(E)_T$ dependences shown in Figs. 1 and 2 and, perhaps also, the two different slopes visible in the curves in Fig. 2. These two different slopes might also be correlated with the two different binding energies of the electrons in the F_1 - and F_2 -centres.

REFERENCES

- 1 K. Biedrzycki, *Phys. Status Solidi (a)* **93**, 503 (1986).
- 2 P.F. Bräunlich, US Patent 3.840.748 (1974).
- 3 V.S. Kortov and R.I. Mints, *Sov. Phys. - Solid State* **9**, 1436 (1967).
- 4 J.J. Tomashpolsky, J.O. Boikova, G.I. Rozenman and M.A. Sievostianov, *Sov. Phys. - Solid State* **20**, 3491 (1978).
- 5 G.I. Rozenman and J.I. Boikova, *Sov. Phys. - Solid State* **21**, 1988 (1979); *Physica Status Solidi (a)* **58**, 379 (1980).

H. GUNDEL et al.

- 6 G.I. Rozenman, I.S. Riez, Yu.L. Chepelev, H.B. Angert and A.A. Zchushkov, *Sov. Phys. - Solid State* **22**, 3466 (1980).
- 7 G.I. Rozenman, I.S. Riez, Yu.L. Chepelev and H.B. Angert, *Zh. Tekh. Fiz.* **51**, 404 (1981).
- 8 G.I. Rozenman, V.A. Okhapkin, Yu.L. Chepelev and V.Ya. Shur, *Zh. Exp. & Teor. Fiz.* **39**, 397 (1984).
- 9 H. Gundel, J. Handerek, H. Riege, E.J.N. Wilson and K. Zioutas, preprint CERN/PS 89-35 (AR), paper submitted to *Ferroelectrics*.
- 10 G.H. Heartling and C.E. Land, *J. Am. Ceram. Soc.* **54**, 1 (1971).
- 11 J. Handerek, J. Kwapulinski, Z. Ujma and K. Roleder, *Ferroelectrics* **81**, 253 (1988).
- 12 Z. Ujma and J. Handerek, *Phase Transitions* **1**, 363 (1980).